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Radiochemical Analysis for Nuclear Waste Management in Decommissioning

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Abstract

The NKS-B RadWaste project was launched from June 2009. The on-going decommisioning activities in Nordic coutries and current requirements and problems on the radiochemical analysis of decommissioning waste were discussed and overviewed. The radiochemical analytical methods used for determination of various radionculides in nuclear waste are reviewed, a book was written by the project partners Jukka Lehto and Xiaolin Hou on the chemistry and analysis of radionuclide to be published in 2010. A summary of the methods developed in Nordic laboratories is described in this report. The progresses on the development and optimization of analytical method in the Nordic labs under this project are presented

Key words

Radioanalysis, radionculides, decommissioning, radioactive waste

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NKS-B RadWaste project report 2010

Edited by ***Xiaolin Hou***

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Content

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1. Introduction

Since a large number of research and power reactors were built in 1950's -1960's and 1960's-1970's respectively, worldwide some 100 power reactors, 250 research reactors, and many other nuclear facilities are or will be decommissioned (IAEA, 2009). In Nordic countries, some nuclear reactors and nuclear facilities have been closed and a few of them have been decommissioned, while many of them are going to be decommissioned (Andersson et al. 2006). In Denmark, three research reactors have been shut down, two of them (DR1 and DR2) have been decommissioned until 2008, and the third reactor (DR3) and a hot cell facility are going to be decommissioned (DD, 2009). In Sweden, up to 2007, 6 nuclear reactors have been shut down, of them only one (R1 at Royal Institute of Technology) was decommissioned, and other 5 (two research reactors in Studsvik R2 and R2-0, power reactors in Ågesta, and Barsebäck 1 and 2) are going to be decommissioned (SKB 2007). In Norway, two small research reactors (JEEP I and NORA) were shut down in 1960's and have been decommissioned to stage 2 and 3 (NEA 2009), while two research reactors (JEEP II and HBWR), which started to operate in 1960's, are going to be shut down and decommissioned sooner or later.

In decommissioning of nuclear facilities, the radioactivity of various radionuclides has to be measured for estimation of the total inventory of radioactivity and its variation with time. The results of measurement are important for categorising the produced waste. In nuclear reactor, besides the nuclear fuel, various materials close to the reactor core are classified as high level radioactive waste. Graphite and biological shield concrete make up considerable low-medium radioactive waste. While other construction materials and used materials in the reactor, such as water, resin are also required to be measured although the volume of these materials may not be a main contribution to the total waste. In addition, the environmental samples surrounding the nuclear reactor and power plant are also required to be analyzed to evaluate the contamination level for possible decontamination. The radioactivity in these materials comes from many nuclides, primarily ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{60}Co, ^{55}Fe, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{129}I, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{154}Eu, and some transuranics (Evans et al. 1988, IAEA 1998, 2007). While in other nuclear facilities, such as hot cell facilities, spent nuclear fuel reprocessing facilities, transuranics are the most important radionuclides (IAEA 2007). Of these radionuclides, gamma emitters such as ^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu, and ^{154}Eu can be instrumentally determined by gamma spectrometry. However, the beta and alpha emitters have to be chemical separated from the matrix and other radionuclides before measurement. Table 1 lists the most important radionuclides for decommissioning of nuclear facilities and their properties. Most of the beta activity is contributed from ^{14}C, ^{3}H, ^{55}Fe, ^{63}Ni, and ^{41}Ca, especially in the first few decades after the final shut down of the reactor. Due to high mobility and long half-life of ^{36}Cl, ^{99}Tc and ^{129}I, and high radiation toxicity of transuranics, the determination of

these radionuclides in radioactive waste is also important for decommissioning and disposal of the waste. ^{14}C, ^{3}H, ^{36}Cl, ^{63}Ni, ^{99}Tc and ^{129}I are pure beta emitters; except ^{90}Sr, others are low energy beta emitters. ^{41}Ca and ^{55}Fe decay by electron capture, while, most of transuranics are alpha emitters. It is therefore necessary to decompose samples and separate individual radionuclides from matrix elements and other interfering nuclides before measurement of their radioactivity. Although much effort has been carried out, the accurate determination of pure beta radionuclides by radiochemical methods is still a major challenge in the characterization of the decommissioning waste due to the complex matrix components of various decommissioning waste, high requirement for the complete radiochemical separation of radionuclides and the relatively low concentrations of most of these radionuclides compared with the gamma-beta emitting radionuclides, especially at the time of decommissioning.

Table 1 The most important radionuclides for decommissioning of nuclear facility

Nuclides	Half life	Decay	Production	Measurement
^{3}H	12.33 y	β	Neutron activation of ^{2}H, Li	LSC
^{14}C	5730 y	β	Neutron activation of ^{14}N, ^{13}C, ^{17}O	LSC
^{36}Cl	3.01×10^{5} y	β	Neutron activation of ^{35}Cl	LSC
^{41}Ca	1.03×10^{5} y	EC	Neutron activation of ^{40}Ca	LSC, TIMS
^{55}Fe	2.73 y	EC	Neutron activation of ^{54}Fe, ^{56}Fe	LSC
^{63}Ni	100 y	β	Neutron activation of ^{62}Ni, ^{63}Cu	LSC, AMS
^{59}Ni	7.6×10^{4} y	EC	Neutron activation of ^{58}Ni	LSC. AMS
^{90}Sr	28.8 y	β	Fission product	LSC, β counting
^{94}Nb	2.03×10^{4} y	β^{-}, γ	Fission product	LSC, γ-spectrometry
^{99}Tc	2.11×10^{5} y	β-	Fission product	LSC, ICP-MS
^{129}I	1.57×10^{6} y	β-	Fission product	LSC, β counting NAA
^{93}Zr	1.53×10^{6} y	β-	Neutron activation of ^{92}Zr	LSC
^{93}Mo	4.0×10^{5} y	EC	Neutron activation of ^{92}No	LSC, AMS
^{239}Pu	2.4×10^{4} y	α	Neutron capture of ^{238}U	α–spectrometry, ICP-MS
^{240}Pu	6.56×10^{3} y	α	Neutron capture of ^{239}Pu	α–spectrometry, ICP-MS
^{241}Pu	14.35	β-	Neutron capture of ^{240}Pu	LSC
^{241}Am	432.2 y	α	Decay from ^{241}Pu	α–spectrometry
^{237}Np	2.14×10^{6} y	α	Neutron capture of ^{238}U	α–spectrometry, ICP-MS
^{243}Cm	29.1 y	α	Neutron capture and decay	α-spectrometry

For the decommissioning of nuclear facilities and characterization of waste, some radiochemical analytical methods have been developed in Nordic laboratories for the determination of these radionuclides in some types of samples, such as graphite and concrete (Hou, 2005a, b, c, 2007; Holm, 1993; Koivula et al. 2003; Lehto & Hou 2009, Sidhu 2006, Skwarzec et al. 2001; Persson, 2005; Persson et al. 2000; Magnusson et al. 2008). However, since there are many different type of nuclear waste from decommissioning activities, the reported methods do not cover all types of the waste, there are always requirement to develop new methods for different types of samples. This project was launched in June 2009, aiming at to establish a close collaboration among the Nordic labs to exchange the experience, overview the analytical methods and capacities in Nordic countries, to develop and improve the methods for the radiochemical analysis of nuclear waste and to meet the gaps and requirement from the nuclear industries, especially decommissioning companies by gathering partners from both Nordic radiochemical analysis laboratories and industries involving in nuclear power and decommissioning of nuclear facilities. This report summarized the activities carried out in this project.

2. Overview of the analytical activities and capacities in the Nordic labs (project partners' labs)

In the project partners labs, including Risø-DTU, Helsinki University and Institute for Energy and Technology (IFE), many radioanalytical methods have been developed and being applied for the determinaiton of radionuclides in radioactive waste from nuclear facilities and surrounding environment, as well as from the decommissioning of nuclear facilities. An overview of these activities are summarized here.

2.1 Analytical activity and capacity in Risø-DTU, Denmark

In the Department of Radiation Research, Risø-DTU, series of radioanalytical methods have been developed, updated and optimized and been applied successfully for the determination of radionuclides, including ^{3}H, ^{90}Sr, ^{99}Tc, ^{129}I, ^{137}Cs, ^{134}Cs, ^{237}Np, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am and ^{244}Cm in various environmental samples including water, soil, sediment, vegetations, animal tissues, and aerosol. Since 2002, with the permanent closing of Risø's nuclear research reactors, and the requirement for characterising decommissioning materials from the reactors as well as their environments, a number of methods for the determination of various radionuclides that are difficult to measure, including ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe and ^{63}Ni, in various samples, e.g. water, soil/sand, concrete, organic materials,

vegetation, graphite, metals, oils and paint have been developed. The methods previously developed for environmental samples have been optimized for the determination of ^{90}Sr, ^{99}Tc, ^{129}I, ^{137}Cs, ^{134}Cs, ^{237}Np, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am and ^{244}Cm in various samples from decommissioning of nuclear reactors.

Since 2002, Risø DTU has been involved in the analysis of various decommissioning materials from Danish research reactors DR1, DR2 and DR3 and other nuclear facilities (such as hot cells and waste station) at Risø site with collaboration with Danish Decommissioning for the determination of radionuclides that are difficult to measure (all beta and alpha emitters and some low level gamma emitters), including ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{129}I, ^{237}Np, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am and ^{244}Cm. Various types of materials have been analyzed for this purpose, including soil/sand, sediment/slurry, concrete, graphite, ordinary water, heavy water, metals/alloys, silicate gels, paint, oil etc. The analytical results have been used to characterize these materials and treatment of the produced wastes.

Risø-DTU has also performed analyses of bio-shielding concrete samples from the Ågesta reactor in Sweden for the determination of ^{3}H, ^{14}C, ^{55}Fe and ^{63}Ni. The analytical results were used for the characterization of the bio-shielding materials in this reactor. Since 2006, a number of samples from Studsvik Nuclear AB, Sweden, including, oil, hydrate, sand, and metals have been analysed for the determination of ^{3}H, ^{14}C, ^{55}Fe and ^{63}Ni. The results were used for the characterization of different nuclear wastes.

A research reactor (100 kW) of the Australian Nuclear Science & Technology Organisation is being decommissioned. In 2009-2010, Risø-DTU carried out analysis of concrete and graphite samples from the decommissioning of this reactor for the radionuclides ^{3}H, ^{14}C, ^{36}Cl, ^{55}Fe, ^{41}Ca and ^{63}Ni. The analytical results have been used for the characterization of these materials.

The Ignalina power plant in Lithuania has two RBMK-1500 units, with total power of 2700 MW, and started operation in 1984. The unit 1 was closed in 2004 and the unit 2 closed in 2009. Although formal decommissioning of the power plant has not yet started, some analyses and characterization of materials has been carried out. Risø-DTU has collaborated with the Institute of Physics, Vilnius, for the analysis of different types of samples including soil, sediment, vegetation, and dust for ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{63}Ni, ^{129}I, Pu isotopes, and ^{241}Am.

Analytical capacity of Risø-DTU for radioactive waste and contaminated environmental samples are summarized in Table 2.

Table 2 Analytical methods and capacity for radioactive waste and contaminated environmental samples

Sample type	Nuclides *	Separation method **	Measurement method	Sample amount (g)	Sample form	Detection limit (Bq/g)	Analytical capacity
Concrete, soil, sediment, sand, brick, stones, rock,	^{3}H	(1) Oxidizing combustion	LSC	2-5 g	Power (<2 mm)	0.1 Bq/g	50 samples/day/ person
	^{14}C	(1) Oxidizing combustion	LSC			0.05 Bq/g	
	^{55}Fe	(2) precipitation-chromatography	LSC	2-20 grams (depend on the activity level)		0.03 Bq/g	10 samples /week/person
	^{63}Ni	(2) precipitation-chromatography	LSC			0.01 Bq/g	
	^{36}Cl	(3) precipitation-chromatography	LSC			0.01 Bq/g	
	^{129}I	(3) Solvent extraction	LSC/AMS			0.01 Bq7g	
	^{41}Ca	(4) Precipitation	LSC			0.15 Bq/g	
	^{90}Sr	(5) Precipitation-chromatography	LSC			0.01 Bq/g	
	^{226}Ra	Co-precipitation	LSC/γ-spectrometry			0.01 Bq/g	
	^{210}Po	(8) no chemical separation	α-spectrometry			0.01 Bq/g	
	^{99}Tc	(6) extraction-chromatography	Beta counter /ICP-MS			0.01 Bq/g	
	Pu	(7) chromatography	α-spectrometry /ICP-MS	5-30 g (depending on the activity level)		0.001 Bq/g	30 samples /week/person
	^{237}Np	(7, 8) Chromatography	α-spectrometry /ICP-MS			0.001 Bq/g	
	Am	(8) chromatography	α-spectrometry			0.001 Bq/g	8 samples /week/person
	Cm	(8) chromatography	α-spectrometry			0.001 Bq/g	
	U isotopes	(8) chromatography	α-spectrometry /ICP-MS			10^{-7} Bq/g for ^{238}U	30 samples /week/person
	Th isotopes	(8) chromatograph	α-spectrometry /ICP-MS			10^{-6} Bq/h ^{232}Th	

* All gamma emitters can be directly measured by gamma spectrometry, which are not listed here, but we can do it easily. The detection limit for gamma emitters is about 1-10 Bq/kg depending on the sample size and the nuclide.

Table 2 Analytical methods and capacity for radioactive waste and contaminated environmental samples (Continued)

Sample type	Nuclides *	Separation method **	Measurement method	Sample amount (g)	Sample form	Detection limit (Bq/g)	Analytical capacity
Exchange resin	^{3}H	(1) Oxidizing combustion	LSC	1-3 g	Just it	0.1 Bq/g	50 samples /day/person
	^{14}C	(1) Oxidizing combustion	LSC			0.05 Bq/g	
	^{55}Fe	(2) precipitation-chromatography	LSC	2-10 grams (depend on the activity level)		0.03 Bq/g	10 samples /week/person
	^{63}Ni	(2) precipitation-chromatography	LSC			0.01 Bq/g	
	^{36}Cl	(3) precipitation-chromatography	LSC			0.01 Bq/g	
	^{129}I	(3) Solvent extraction	LSC/AMS			0.01 Bq7g	
	^{41}Ca	(4) Precipitation	LSC			0.01 Bq/g	
	^{90}Sr	(5) Precipitation-chromatography	LSC			0.01 Bq/g	
	^{226}Ra	Co-precipitation	LSC/γ-spectrometry			0.15 Bq/g	
	^{210}Po	(8) no chemical separation	α-spectrometry			0.01 Bq/g	
	^{99}Tc	(6) extraction-chromatography	Beta counter /ICP-MS			0.01 Bq/g	
	Pu	(7) chromatography	α-spectrometry /ICP-MS	2-10 g (depending on the activity level)		0.001 Bq/g	30 samples /week/person
	^{237}Np	(7, 8) Chromatography	α-spectrometry /ICP-MS			0.001 Bq/g	
	Am	(8) chromatography	α-spectrometry			0.001 Bq/g	8 samples /week/person
	Cm	(8) chromatography	α-spectrometry			0.001 Bq/g	
	U isotopes	(8) chromatography	α-spectrometry /ICP-MS			10^{-7} Bq/g for ^{238}U	30samples /week/person
	Th isotopes	(8) chromatograph	α-spectrometry /ICP-MS			10^{-6} Bq/h ^{232}Th	

* All gamma emitters can be directly measured by gamma spectrometry, which are not listed here, but we can do it easily. The detection limit for gamma emitters is about 1-10 Bq/kg depending on the sample size and the nuclide.

Table 2 Analytical methods and capacity for radioactive waste and contaminated environmental samples (Continued)

Sample type	Nuclides *	Separation method **	Measurement method	Sample amount (g)	Sample form	Detection limit (Bq/g)	Analytical capacity
Vegetation, animal tissues (dry mass), wood, paint, bitumen	^{3}H	(1) Oxidizing combustion	LSC	2-5 g	Power (<2 mm)	0.1 Bq/g	50 samples /day/person
	^{14}C	(1) Oxidizing combustion	LSC			0.05 Bq/g	
	^{55}Fe	(2) precipitation-chromatography	LSC	2-20 grams (depend on the activity level)		0.03 Bq/g	10 samples /week/person
	^{63}Ni	(2) precipitation-chromatography	LSC			0.01 Bq/g	
	^{36}Cl	(3) precipitation-chromatography	LSC			0.01 Bq/g	
	^{129}I	(3) Solvent extraction	LSC/AMS			0.01 Bq7g	
	^{41}Ca	(4) Precipitation	LSC			0.15 Bq/g	
	^{90}Sr	(5) Precipitation-chromatography	LSC			0.01Bq/g	
	^{226}Ra	Co-precipitation	LSC/γ-spectrometry			0.01 Bq/g	
	^{210}Po	(8) no chemical separation	α-spectrometry			0.01 Bq/g	
	^{99}Tc	(6) extraction-chromatography	LSC/ICP-MS			0.01 Bq/g	
	Pu	(7) chromatography	α-spectrometry /ICP-MS	5-30 g (depending on the activity level)		0.001 Bq/g	30 samples /week/person
	^{237}Np	(7, 8) Chromatography	α-spectrometry /ICP-MS			0.001 Bq/g	
	Am	(8) chromatography	α-spectrometry			0.001 Bq/g	8 samples /week/person
	Cm	(8) chromatography	α-spectrometry			0.001 Bq/g	
	U isotopes	(8) chromatography	α-spectrometry /ICP-MS			10^{-7} Bq/g for ^{238}U	30samples /week/person
	Th isotopes	(8) chromatograph	α-spectrometry /ICP-MS			10^{-6} Bq/h ^{232}Th	

* All gamma emitters can be directly measured by gamma spectrometry, which are not listed here, but we can do it easily. The detection limit for gamma emitters is about 1-10 Bq/kg depending on the sample size and the nuclide.

Table 2 Analytical methods and capacity for radioactive waste and contaminated environmental samples (Continued)

Sample type	Nuclides *	Separation method **	Measurement method	Sample amount (g)	Sample form	Detection limit (Bq/g)	Analytical capacity
Metals (aluminum, stainless steel, and others)	^{3}H	(1) Oxidizing combustion/ digestion	LSC	2-5 g	Small pieces (<2 mm)	0.1 Bq/g	50 samples /day/person
	^{14}C	(1) Oxidizing combustion/digestion	LSC			0.05 Bq/g	
	^{55}Fe	(2) precipitation-chromatography	LSC	2-20 grams (depend on the activity level)	Pieces (>10 mm in diameter)	0.03 Bq/g	10 samples /week/person
	^{63}Ni	(2) precipitation-chromatography	LSC			0.01 Bq/g	
	^{36}Cl	(3) precipitation-chromatography	LSC			0.01 Bq/g	
	^{129}I	(3) Solvent extraction	LSC/AMS			0.01 Bq7g	
	^{41}Ca	(4) Precipitation	LSC			0.15 Bq/g	
	^{90}Sr	(5) Precipitation-chromatography	LSC			0.01 Bq/g	
	^{226}Ra	Co-precipitation	LSC/γ-spectrometry			0.01 bq/g	
	^{210}Po	no chemical separation	α-spectrometry			0.01 bq/g	
	^{99}Tc	(6) extraction-chromatography	LSC/ICP-MS			0.01 Bq/g	
	Pu	(7) chromatography	α-spectrometry /ICP-MS	5-30 g (depending on the activity level)		0.001 Bq/g	30 samples /week/person
	^{237}Np	(7, 8) Chromatography	α-spectrometry /ICP-MS			0.001 Bq/g	
	Am	(8) chromatography	α-spectrometry			0.001 Bq/g	8 samples /week/person
	Cm	(8) chromatography	α-spectrometry			0.001 Bq/g	
	U isotopes	(8) chromatography	α-spectrometry /ICP-MS			10^{-7} Bq/g for ^{238}U	30samples /week/person
	Th isotopes	(8) chromatograph	α-spectrometry /ICP-MS			10^{-6} Bq/h ^{232}Th	
	Th isotopes	(8) chromatograph	α-spectrometry /ICP-MS			10^{-6} Bq/h ^{232}Th	

Table 2 Analytical methods and capacity for radioactive waste and contaminated environmental samples (Continued)

Sample type	Nuclides *	Separation method **	Measurement method	Sample amount (g)	Sample form	Detection limit (Bq/g)	Analytical capacity
water	^{3}H	(1) No/electrolysis enrichment/oxidizing combustion	LSC	10-1000 grams (depending on the activity level, for activity higher than 1Bq/g for each radionuclide, 50 g ok for analysis of all radionuclides)	liquid	0.1 Bq/g	50 samples /day/person
	^{14}C	(1) No/electrolysis enrichment/oxidizing combustion	LSC			0.05 Bq/g	
	^{55}Fe	(2) precipitation-chromatography	LSC			0.03 Bq/g	10 samples /week/person
	^{63}Ni	(2) precipitation-chromatography	LSC			0.01 Bq/g	
	^{36}Cl	(3) precipitation-chromatography	LSC			0.01 Bq/g	
	^{129}I	(3) Solvent extraction	LSC/AMS			0.01 Bq7g	
	^{41}Ca	(4) Precipitation	LSC			0.15 Bq/g	
	^{90}Sr	(5) Precipitation-chromatography	LSC			0.01 Bq/g	
	^{226}Ra	Co-precipitation	LSC/γ-spectrometry			0.01 Bq/g	
	^{210}Po	no chemical separation	α-spectrometry			0.01 Bq/g	
	^{99}Tc	(6) extraction-chromatography	LSC/ICP-MS			0.01 Bq/g	
	Pu	(7) chromatography	α-spectrometry /ICP-MS			0.001 Bq/g	30 samples /week/person
	^{237}Np	(7, 8) Chromatography	α-spectrometry /ICP-MS			0.001 Bq/g	
	Am	(8) chromatography	α-spectrometry			0.001 Bq/g	8 samples /week/person
	Cm	(8) chromatography	α-spectrometry			0.001 Bq/g	
	U isotopes	(8) chromatography	α-spectrometry /ICP-MS			10^{-7} Bq/g for ^{238}U	30samples /week/person
	Th isotopes	(8) chromatograph	α-spectrometry /ICP-MS			10^{-6} Bq/h ^{232}Th	

* All gamma emitters can be directly measured by gamma spectrometry, which are not listed here, but we can do it easily. The detection limit for gamma emitters is about 1-10 Bq/kg depending on the sample size and the nuclide.

2.2 Analytical activity in the University of Helsinki

In Laboratory of Radiochemistry, University of Helsinki, a numbers of analytical method have been developed for the determination of alpha emitters including ^{238}U, ^{238}Pu, ^{239,240}Pu, ^{241}Am, ^{242}Cm, ^{243,244}Cm and beta emitters including ^{63}Ni, ^{90}Sr, ^{99}Tc, and ^{129}I, these methods have been applied to analysis of primary coolant, concentrated waste water and ion exchange resins from two Finnish nuclear power plants Loviisa NPP (2 PWR reactors) and Olkiluoto NPP (2 BWR reactors). (Puukko & Jaakkola, 1992).

2.3 Analytical capacity in Institute for Energy and Technology(IFE), Norway

IFE has established a number of methods for the determination of natural occurred as well as anthropogenic radionculides, including isotopes of ^{3}H, ^{90}Sr, ^{226}Ra, ^{228}Ra, ^{241}Am, ^{243}Cm and isotopes of U, Th, and Pu. These methods have been successfully used for the analysis of various environmental samples including contaminated areas by discharges from the nuclear facility at IFE. Although no waste samples from decommissioning of nuclear facilities have been analyzed, these methods can be used for this purpose with some modification.

3. The on-going decommisioning activities in Nordic coutries and current requirements to the radiochemical analysis

Some decommissioning work have being carried out in Denmark and Sweden, while in Norway and Finland, since no reactor has been closed yet, no real decommissioning was done although some planning work are made.

3.1 Decommissioning activity in Denmark

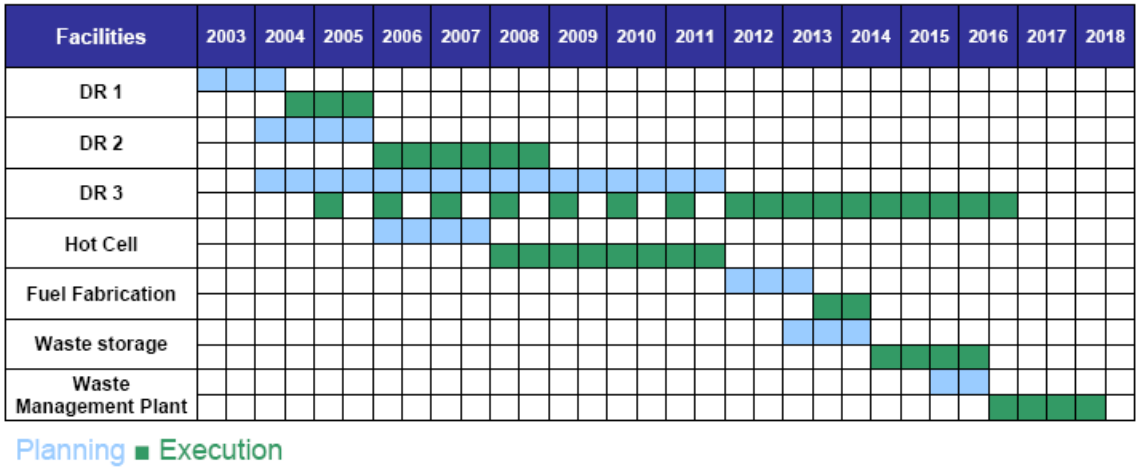
In Denmark, all six nuclear facilities at Risø site are being decommissioned, which include three research reactors, DR1, DR2 and DR3, as well as a Hot Cell facility, a fuel fabrication facility and a waste management plant. All facilities are shut down except the waste management plant, which will be the last of the facilities to be decommissioned, because it is needed during the decommissioning process. DR1 (1957-2001) was a small homogeneous solution-type reactor with a maximum power of 2 kW; it was decommissioned in 2005. DR2 (1959-1975) is a 5 MW light-water reactor of the pool type and was

primarily used for nuclear physical researches and the production of radioactive isotopes. After the closure of this reactor in 1975, it was partly decommissioned in 1980's and completely decommissioned by 2008. DR3 was a heavy water reactor of 10 MW power and neutron flux of up to 10^{14} n/cm^2 s, which operated from 1960 to 2000. DR3 was mainly used for basic neutron physics research, materials researches and the production of radioactive isotopes for medical and industrial use. In addition, neutron irradiation of silicon for the semi-conductor industry was carried out. Some part of this reactor, mainly peripheral system of the reactor hall has being decommissioned since 2005. The major decommissioning will be started from 2012 and to be finished in 2016. The Hot Cell (1964-1989) was used for investigating irradiated fuel and for packaging of radioactive sources for irradiation-facilities. The facility was sealed in 1993 and with decommissioning beginning in 2008. The Fuel fabrication Facility (1961-2000) was used in part for the fabrication of fuel elements for reactor DR3 through the use of enriched uranium powder. The decommissioning of this facility includes removal of various items of inventory and equipment, dismantling or cleaning of the ventilation canals and removal or cleaning of radioactive waste systems, which will start from 2012 and to be finished in 2014. The Waste Management Plant (1958 -) treats the radioactive wastewater from the Risø National Laboratory and Danish Decommissioning. It also receives and treats all radioactive waste from external costumers, such as hospitals and industries as well as from the decommissioning projects. It is responsible for the storage of all radioactive waste, until a decision has been taken concerning the establishment of a Danish final repository for radioactive waste. The Waste Management Plant is still in commission and will continue to be so, until a new and smaller plant is built. The decommissioning of all these facilities is planned to be finished in 2018, Table 3 shows the decommissioning plan of these facilities.

The decommissioning of the reactor core of DR3 is under planning. Different options for decommissioning have been discussed with the international expert panel meeting in January 2010. It was decided to remove and deposit the Top Shield Plug (TSP) and the Top Shield Ring (TSR) as whole units. This will created odd sized units different from the standard containers. The reactor will after removal of TSP and TSR be decommissioned from top to bottom and from inside to outside. Danish Decommissioning has identified a number of ways to complete this task and is in the middle of a process of describing and comparing the different scenarios against each other with regard to economy, feasibility and health physics aspects. An important aspect in the planning of the decommissioning of DR3 is the waste management. Plans are made for number of needed containers, subsamples for

analysis, drying of wet waste, sizes and way to cut the waste item in order to minimize waste volume. Furthermore it is taken into consideration to minimize generation of secondary waste, e.g., new tools or plastics covers used as equipment during the decommissioning process. From 2009 to 2011 more free space is created in the reactor building by taking down the peripheral systems of DR3. Many of the waste items created this way are released as conventional waste.

Table 3 Decommissioning plan for nuclear facilities in Denmark (http://www.ddcom.dk/)



3.2 Decommissioning activities in Sweden

In Sweden, up to 2007, 6 nuclear reactors have been shut down, of them only one (R1 at Royal Institute of Technology) was decommissioned, and other 5 (two research reactors in Studsvik R2 and R2-0, power reactors in Ågesta, and Barsbäck 1 and 2) are going to be decommissioned (SKB 2007).

Both Barsebäck One and Two nuclear power plant units are boiling water reactors, that were in operation from1977-1999 and 1977-2005, each with a thermal power of 1815 MW. The fuel elements in the plants have been removed and a substantial system decontamination at both units has been carried out.

In the reactor tank and pool water, some uranium and transuranics are still left, which is attributed to the neutron detectors which contain some high enriched ^{235}U. A characterization of soil, groundwater and sediment in the surrounding area of the plant is going to be carried out in 2010-2011 for long-lived radionuclides. The decommissioning of the reactors is planned to begin by 2020, while the characterization of the construction materials, the estimation of the radioactivity inventory will be

carried out before 2020. The measurement of some radionuclides as vectors with relative short half life are important at the moment, otherwise they will decay away when decommissioning is carried out after 10 years.

Two research reactors R2 (5 MW) and R2-0 (1 MW) were commissioned in 1960 and 1969. On June 16th 2005, the operation of the R2 and R2-0 reactors ceased and the reactors were closed. Ågesta nuclear power plant was the first Swedish commercial nuclear power plant. The R3 Ågesta reactor (10 MW), was a natural uranium reactor and operated from 1964 to 1974. SVAFO will be responsible for decommissioning the reactors in Ågesta and Studsvik (R2 and R2-0). This work is expected to be completed by 2045.

3.3 Requirements to radiochemical analysis during decommissioning

During the decommissioning of Danish nuclear facilities, the most often measured radionculides are ^{3}H, ^{14}C, ^{55}Fe, and ^{63}Ni, although many other radionculides including gamma, beta and alpha emitters were also required to be measured. However, the challenge for radiochemical analysis required for decommissioning is the analysis of many different type of materials, some of which are difficult to be decomposed or the conventional analytical method cannot be applied. For example, during decommission of hot cell, it was found that most of radioactivity is from the surface materials made of PCBs in the inside of the hot cell. Since PCB is resistant to heat, acid and alkali, it is very difficult to be decomposed. Another example is analysis of aluminum materials for ^{3}H and ^{14}C. A combustion method using Sample Oxidizer has been developed and used at Risø-DTU for determination of ^{3}H and ^{14}C in many solid samples. However, aluminum can react with platinum during combustion in the Oxidizer, which will damage the combustion basket made of platinum, and make the method invalid. In addition, the species of radionuclides, such as free tritium (water associated) and bound tritium (in crystal of concrete) in concrete, was required during the estimation of ^{3}H in the biological concrete shielding in Danish reactor DR3. This requires the development of a new analytical method for speciation of tritium in concrete.

Besides the radionuclides which are often required, some long-lived and low concentration radionuclides were also required. For example, the characterization of environment in Barsebäck NPP requires the determination ^{79}Se, ^{94}Nb, ^{93}Mo, ^{93}Zr, ^{126}Sn. Since the methods in the literature for these

radionuclides mainly focus on the analysis of nuclear fuel solution. New methods have to be developed to meet this requirement.

4. Overview of the radiochemical analytical methods for radionuclides difficult to measure in nuclear waste

Many methods have been reported in the literature, a comprehensive review of radiochemical analytical methods for determination of long-lived radionuclides in the waste from decommissioning of nuclides facilities as well as in environmental samples has been prepared as a book (Lehto and Hou 2010) by the participants of this project.

In Nordic labs, a series of methods have been published in journal articles and scientific reports. Some of these documents are listed below and the methods for the radionuclides concerned are briefly described.

List of published articles and scientific reports by our laboratory for the determination of radionuclides for environmental and decommissioning waste samples:

R1: *Xiaolin Hou*, Determination of ^{14}C and ^{3}H in Reactor Graphite and Concrete for Decommission, *Applied Radiation and Isotopes*, 62(2005)871-882

R2: *Xiaolin Hou*, Determination of ^{63}Ni and ^{55}Fe in nuclear waste and environmental samples, *Anal. Chim. Acta*, 53582005)297-307

R3: *Xiaolin Hou, Lars Frøsig Østergaard, Sven P. Nielsen*, Determination of ^{36}Cl in Nuclear Waste from Reactor Decommissioning, *Anal. Chem*, 79(2007), 3126-3134.

R4: *Xiaolin Hou*, Radiochemical determination of ^{41}Ca in reactor concrete for decommissioning, *Radiochim., Acta,* 93(2005)611-617

R5: *Q.J.Chen, X.L.Hou, S.P. Nielsen, H. Dahlgaard,Y.Yu*, Separation of Sr from Ca, Ba and Ra in the determination of radiostrontium by means of simple precipitation using $Ca(OH)_2$, $Ba(Ra)Cl_2$ or $Ba(Ra)SO_4$. *Anal. Chim. Actc*, 466 (2002)109-116

R6: *Qiao Jixin, Hou Xiaolin, Roos Per, Miró Manuel*, (2009), Rapid Determination of Plutonium Isotopes in Environmental Samples Using Sequential Injection Extraction Chromatography and Detection by Inductively Coupled Plasma Mass Spectrometry, *Anal. Chem*., 81(2009)8185-8192

R7: Keith-Roach, M.J.; Stürup, S.; Oughton, D.H.; Dahlgaard, H., Comparison of two ICP-MS set-ups for measuring ^{99}Tc in large volume water samples. *Analyst* (2002) **127** , 70-75

R8: Hou, X.L.; Dahlgaard, H.; Rietz, B.; Jacobsen, U.; Nielsen, S.P.; Aarkrog, A., Determination of ^{129}I in seawater and some environmental materials by neutron activation analysis. *Analyst* (1999) **124** , 1109-1114

R9: Chen, Q.J.; Nielsen, S.P.; Aarkrog, A.; Dahlgaard, H.; Duniec, S., Distribution of plutonium isotopes in cooling water from a PWR. *J. Nucl. Sci. Technol.* (1993) **30** , 164-170

R10: Q.J. Chen, A. Aarkrog, S.P. Nielsen, H. Dahlgaard, B. Lind, A.K. Kolstad, Y.X. Yu, Procedures for determinaton of Pu, Am, Np, U, Th, ^{99}Tc, ^{210}Po-^{210}Po in environmental materials, Risø report, Risø-R-1263, 2001. http://130.226.56.153/rispubl/NUK/nukpdf/ris-r-1263.pdf

R11 Puukko E. and Jaakkola T. (1992). Actinides and beta emitters in the process water and ion exchange resin samples from the Loviisa power plant, report YJT-92-22

R12. Hou X.L. Radiochemical analysis of radionuclides difficult to measure, *J. Radioanal. Nucl. Chem.*, 2007, 273, 43-48.

The analysis of samples for various radionuclides is divided into a few steps: (1) Pre-treatment and preparation of samples; (2) Pre-concentration of target radionuclides from sample matrices; (3) Radiochemical separation and purification of target radionuclide; (4) Measurement of radionuclides; (5) Data analysis and results report.

4.1 Pre-treatment and preparation of samples

4.1.1 Water samples

Water samples are normally filtered through a 0.45 μm filter to remove the particles and suspended material. For some samples, where the particles should be included, no filtration is carried out. For the determination of tritium, the water is distilled after filtration.

4.1.2 Concrete, graphite, metals, exchange resin, soil/sediment samples

Soil/sediment samples are first air dried, with large stones/rocks and roots of plants removed. The dried sample is then crushed, sieved and mixed to obtain a relatively homogeneous sample. Exchange resin

samples are directly used for analysis without drying. Metal samples need to be cut into small pieces before analysis. For the determination of non-volatile radionuclides (i.e. isotopes of Ca, Fe, Ni, Sr, Nb, Pu, Am and Cm), samples are ashed at 450-550 °C overnight. For the determination of ^{3}H, ^{14}C, dried samples are combusted (see separation procedure). For the determination of ^{36}Cl, ^{129}I and ^{99}Tc, samples are either treated by acid digestion in a closed system (Fig. 1), or by an alkali fusion procedure (sample + NaOH, heat at 500-550 °C for sample decomposition, but not for metal samples).

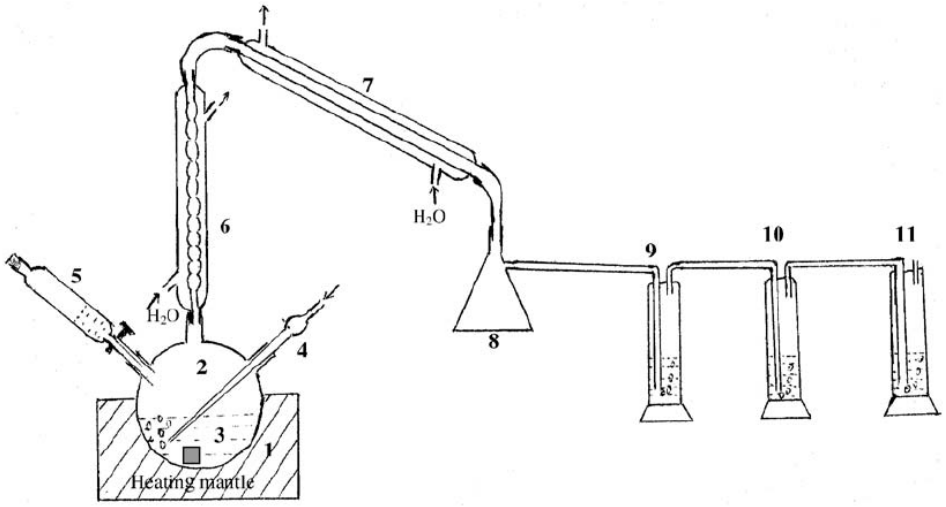


Fig. 1 Schematic diagram of acid digestion system for the decomposition of graphite and concrete
1-Heating mantle; 2-three-neck-flask; 3-sample and digestion acids; 4-bubbling tube; 5-separation funnel for adding acids; 6,7-reflux cooler; 8-condenser receiver; 9-washing bottle with diluted acid; 10, 11-absorption bottle with NaOH

4.1.3 Vegetation/animal tissue samples

Samples are either first air dried (vegetation) or freeze dried (animal tissues), and then ground to a powder. For the determination of non-volatile radionuclides (i.e. isotopes of Ca, Fe, Ni, Sr, Nb, Pu, Am and Cm), the dried samples are ashed at 450-500 °C overnight. For the determination of ^{3}H, ^{14}C, the dried samples are combusted (see separation procedure). For the determination of ^{36}Cl and ^{129}I, the

samples are either treated by acid digestion in a closed system (Fig.3), or by an alkali fusion procedure (sample + NaOH, heat at 500-550 °C for sample decomposition).

4.2 Pre-concentration and separation of nuclides from sample matrices

4.2.1 Solid samples

Ashed samples are digested with acids (*aqua regia*) to release all metals, including transuranics to the solution. To the acid digested samples (solution/leachate), stable Fe, Ni and Nb are added as carrier, and Co, Cu, Eu, Cs, etc. are added as hold-back carrier, whereas ^{99m}Tc, ^{242}Pu and ^{243}Am are added as tracers. The pH of the solution is adjusted to 8-9 with NaOH to precipitate radionuclides of transition elements and transuranics including ^{55}Fe, ^{63}Ni, ^{94}Nb, Pu isotopes, ^{241}Am and ^{244}Cm with $Fe(OH)_3$. After centrifuging, the supernatant is acidified with HNO_3 and the solution is loaded to an anion exchange column to separate ^{99}Tc, the effluent is used for the determination of ^{41}Ca and ^{90}Sr. The precipitate is dissolved with HCl and HNO_3. The remaining precipitate is separated by centrifuge and used for the determination of ^{94}Nb. To the supernatant, NaOH is added to adjust pH 8-9 to separate the precipitation of $Fe(OH)_3$ which is used for the determination of ^{55}Fe, ^{63}Ni, Pu isotopes, ^{241}Am and ^{244}Cm.

To the acid digested samples, ^{36}Cl and ^{129}I are released from the sample and collected in the trap solution and washed off the walls of condenser and tubes. Subsequently, they are separated by AgCl precipitation and solvent extraction respectively.

4.2.2 Water samples

For radionuclides of transition metals and transuranics, the pre-concentration is carried out by co-precipitation with $Fe(OH)_3$. The supernatant is acidified with HNO_3 and the solution is loaded to an anion exchange column to separate ^{99}Tc, and the effluent is used for the determination of ^{41}Ca and ^{90}Sr, which is similar to the acid leaching of the solid sample mentioned above. For ^{129}I, solvent extraction is directly used for the separation of iodine from matrix; the aqueous phase from the extraction is used for separation of ^{36}Cl by AgCl precipitate, while for ^{14}C, an evaporation method is used.

4.3 Radiochemical separation and purification of target radionuclide

4.3.1 Procedure for separation of ^{3}H and ^{14}C in solid samples (concrete, graphite, metals, vegetation, resin, soil/sediment etc.)

A rapid combustion method developed in our laboratory will be used for this analysis (**R1**). The ground sample is first mixed with cellulose powder in a CombustCone, which is put into a Sample Oxidizer. In the Sample Oxidizer, the sample is combusted at 1200°C with oxygen flow. All tritium in the samples is converted to H_2O vapour during combustion with oxygen, which is flushed with nitrogen gas through a closed system (Fig. 2), and condensed in an air condenser, and the condensed tritiated water is collected in a LSC vial after the combustion, the scintillation cocktail stored in a reservoir is used to flush the tubes and combined to the vials with tritiated water. All ^{14}C in the samples including carbon, carbonate and organic carbon are converted to CO_2, and flushed through the system (Fig.2), and finally absorbed in CarboSorb (an amine solution) filled into a column. The absorbed ^{14}C in the solution is collected in a LSC vial after the combustion, and the tube is washed with a scintillation cocktail stored in a reservoir to remove any remaining Carbosorb in the tubes. The separated tritium and ^{14}C in the vials are directly measured by LSC. The whole procedure of combustion takes about 2 minutes.

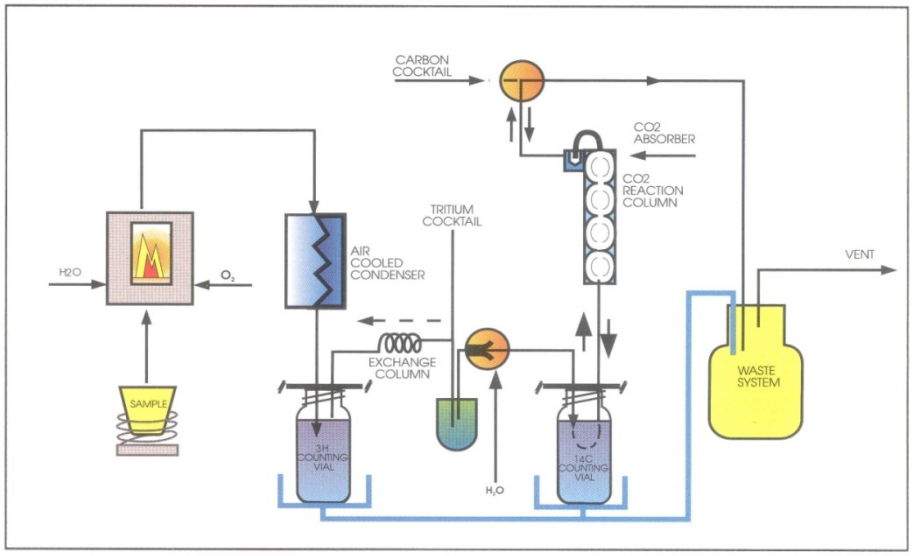


Figure 2. Diagram of sample oxidizer used for separation of tritium and ^{14}C from solid samples by combustion.

4.3.2 Separation procedure for ^{14}C in water samples

The water is taken to a round flask, Na_2CO_3 carrier is added, and pH of the water is adjusted to 7-9, and connected to an evaporation system. A washing bottle filled with Carbonsorb is connected to the

evaporation system, and the washing solution is checked for any volatile ^{14}C species in the water samples. The water is evaporated to 0.2-0.3 ml. The concentrated water is transferred to a Combust Cone filled with cellulose powder. The combustion method described above for the solid samples is used for separation of ^{14}C.

4.3.3 Separation of ^{129}I in water and acid digested samples or alkali ashed samples

After the acid digestion of the sample, the iodine is released from the sample solution and remains in the trap solution and on the condenser walls. The trap solution and washes of the tubes and condenser are combined as iodine trap solution. Solid samples (soil, sediment and vegetation) decomposed by an alkali fusion method are leached with water, and filtered through a filter paper. For water samples, NaOH and NaClO are added, and the sample heated for 2-3 hours to decompose organics in the water. ^{129}I in the filtrate/trap solution and water samples is then separated by solvent extraction (**R8**). 1-2 mg of iodine carrier (^{127}I) is first added to the filtrate (trap solution or digested water), then $KHSO_3$ is added and pH is adjusted to 1-2 using HCl to convert iodine to iodide. Iodine is then extracted using $CHCl_3$ after addition of $NaNO_2$ to oxidize iodide to elemental iodine (I_2). Iodine in the $CHCl_3$ phase is back extracted using $KHSO_3$ solution, this extraction and back-extraction step is repeated to purify ^{129}I. The separated ^{129}I in 3-5 ml of diluted $KHSO_3$ solution is used for LSC measurement of ^{129}I.

4.3.4 Separation of ^{36}Cl in water and digested samples

After acid digestion of the sample, chlorine is released from the sample solution and remains in the water trap solution and on the condenser walls. The trap solution and washes of the tubes and condenser are combined as chlorine trap solution. ^{36}Cl in the solution is then separated by precipitation and ion exchange methods after the separation of iodine using solvent extraction (**R3**). To the aqueous phase from the solvent extraction of iodine, 5-10 mg stable chlorine (^{35}Cl) is added, and then $NaNO_2$ is added to convert all chlorine to chloride. $AgNO_3$ is then added to precipitate chloride as AgCl, which is then separated by centrifuging. The AgCl precipitate is dissolved with NH_4OH and then Cl^- is re-precipitated as AgCl by addition of HNO_3 to pH 1-2. The formed AgCl is dissolved in 5 ml NH_4OH and loaded to an anion exchange column which has been conditioned with NH_4OH solution. After washing with NH_4OH to remove all Ag^+, Cl^- adsorbed on the column is finally eluted with NH_4NO_3-

NH_4OH solution. The eluate is evaporated to dryness and the residue is dissolved with 2-3 ml water and transferred to a vial for LSC measurement of ^{36}Cl.

4.3.5 Separation of ^{99}Tc from digested sample and water

To the supernatant from the precipitation of transition metals and transuranics, HNO_3 is added to adjust pH to 3-5, the solution is then loaded to an anion exchange column (AG1×4, Cl^- form), the effluent is used for the determination of ^{41}Ca and ^{90}Sr. The column is washed with 0.5 M HNO_3, and the ^{99}Tc on the column is finally eluted using 10M HNO_3. The eluate is evaporated to 0.5-1.0 ml on a hotplate, and then diluted with 20 ml H_2O. The prepared solution is loaded to a 2 ml TEVA extraction chromatographic column, the column is washed with 2M HNO_3, ^{99}Tc on the column is finally eluted with 8M HNO_3. The eluate is evaporated to 0.5-1.0 ml and diluted to 5 ml with H_2O. Chemical yield is measured by gamma counting of ^{99m}Tc, and the solution is finally analyzed using ICP-MS for the measurement of ^{99}Tc (**R7**).

4.3.6 Separation of ^{41}Ca and ^{90}Sr

To the effluent from the ion exchange separation of ^{99}Tc, Sr^{2+} and Ca^{2+} carriers, Fe^{3+} hold back carrier and ^{85}Sr tracer are added, and the solution is adjusted to pH 9 using NaOH. After separation of $Fe(OH)_3$ precipitate, Na_2CO_3 solution is added to the supernatant, and the formed precipitate of Ca and Sr as carbonate is separated by centrifuging. The precipitate is dissolved with HCl, and 2 mg of Fe^{3+}, Co^{2+}, and Eu^{3+} are added, and then the NaOH is added to adjust pH to 8-9. The $Fe(OH)_3$ precipitate is removed by centrifuging, 6 M NaOH is added to the supernatant to a final concentration of NaOH to 0.5 M, and the precipitate of $Ca(OH)_2$ is separated by centrifuging. The supernatant is used for the separation of ^{90}Sr. The $Ca(OH)_2$ precipitate is dissolved with HCl, 2mg of Fe^{3+}. Co^{2+} and Eu^{3+} are added and pH adjusted to 8-9 using NaOH. The precipitate is removed by centrifuging, and 6M NaOH is added to the supernatant to 0.5 M of NaOH. $Ca(OH)_2$ precipitate is then separated by centrifuging. This dissolution and precipitation step is repeated. The final obtained $Ca(OH)_2$ precipitate is dissolved in HCl, chemical yield is measured by determination of Ca in the final solution and the solution before separation of Ca. ^{41}Ca in the final solution is measured by LSC counting (**R4**).

To the supernatant from the first $Ca(OH)_2$ precipitation step, HCl is added to adjust pH 2. After addition of Y^{3+} carrier the solution is kept for 3 weeks for in-growth of ^{90}Y. The chemical yield of Sr is

measured by gamma counting of ^{85}Sr in the solution. Afterwards, Na_2SO_4 solution is added to precipitate Sr as well as Ra, and Ba as sulphate. To the supernatant, NH_4OH is added to adjust pH to 8-9, and the precipitate of $Y(OH)_3$ is separated by centrifuging. The precipitate is then dissolved with HCl, $H_2C_2O_4$ is added to the solution to precipitate Y as $Y_2(C_2O_4)_3$, and the chemical yield of Y is measured by weighing the dried $Y_2(C_2O_4)_3$ precipitate. ^{90}Y in the precipitate is measured by beta counting to determine ^{90}Sr in the samples with correction for chemical yield of Sr and Y, as well as the in-growth time of ^{90}Y from ^{90}Sr (**R5**).

4.3.7 Separation of ^{55}Fe and ^{63}Ni

The precipitate of transition metals and transuranics in $Fe(OH)_3$ is dissolved with HCl, and divided into two aliquots, one for transuranics. To another aliquot, concentrated HCl is added to 9 M HCl, this solution is loaded to an anion exchange column (AG1×4, Cl^- form), and the effluent is collected for ^{63}Ni determination. The column is washed with 4 M HCl to remove Co^{2+}. Cu^{2+} and other transition metals, and ^{55}Fe on the column is eluted with 0.5 M HCl. The eluate is evaporated to dryness, and the residue is dissolved with 9M HCl. The solution is loaded to a new anion exchange column. The column is washed with 4M HCl, and ^{55}Fe on the column is finally eluted with 0.5 M HCl. The eluate is evaporated to dryness, and dissolved with 1M H_3PO_4. The Fe in the final solution and the original solution is measured by ICP-OES to calculate the chemical yield. The remaining solution is used to measure ^{55}Fe by LSC (**R2**).

The effluent from the anion exchange column is evaporated to dryness, the residue is dissolved with 1M HCl, and 1ml of 1M sodium citrate is added to the solution. Then NH_4OH is added to the solution to pH 8-9. The solution is then loaded to an extraction chromatographic column (Eichrom Ni-column, 2 ml), the column is washed with 0.2 M sodium citrate in pH 9, and Ni on the column is finally eluted using 3M HNO_3. The eluate is evaporated to 0.1-0.3 ml on a hotplate, and transferred to a vial with 2 ml water. 0.1 ml of the final solution, as well as the original solution, are taken for the determination of Ni using ICP-OES for the chemical yield measurement. ^{63}Ni in the remaining solution is measured by LSC (**R2**).

4.3.8 Separation of Pu isotopes

To the aliquot of dissolved hydroxides precipitation, $KHSO_3$ solution is added to reduce Pu to Pu(III), then NaOH is added to adjust pH to 8-9 and the precipitate formed is separated by centrifuging. The precipitate is dissolved with concentrated HCl, and then the HNO_3 is added to a concentration of 8M. The solution is loaded to an anion exchange column (AG1×4, Cl^- form), and the effluent is collected for the determination of ^{241}Am and ^{244}Cm. The column is washed with 8M HNO_3, and 9M HCl, Pu on the column is finally eluted using 0.05M $NH_2OH{\cdot}HCl$-2M HCl solution. The eluate is evaporated to dryness; the residue is dissolved with 0.5m HNO_3. Pu isotopes in the solution are measured by ICP-MS or alpha spectrometry after electrodeposition. ^{242}Pu is used for chemical yield monitoring (**R9, R10, R11**).

4.3.9. Separation of Am and Cm

To the effluent from the ion exchange separation of Pu, Fe^{3+} is added, and NH_3 is added to pH 8-9. The precipitate isolated by centrifuging is dissolved in 1.5 ml of concentrated HNO_3, 0.5 ml of 30% H_2O_2 is added and the solution stirred for 2 minutes. 25 ml of CH_3OH is added to the solution to make 93% CH_3OH solution. The solution is loaded to an anion exchange column (AG1×4, Cl^- form), and the column washed using 20 ml of 1m HNO_3-93%CH_3OH and 100 ml of 0.1M HCL-0.5M NH_4SCN-80% CH_3OH, followed by 50ml 1M HNO_3-93% CH_3OH. Am and Cm on the column are finally eluted using 50ml of 1.5M HCl-86%CH_3OH solution. The solution is evaporated to dryness; concentrated HNO_3 is added to the residue and digested for 30 min on a hotplate. The solution is evaporated to dryness. The residue is then dissolved with 10 ml of 0.05M H_2SO_4, and the solution is transferred to an electrodeposition cell, where NH_3 is added to adjust pH to 2.5. Am and Cm are then electrodeposited on a disc, and ^{241}Am and ^{244}Cm on the disc are finally measured by alpha spectrometry. ^{243}Am on the disc is also measured by alpha spectrometry, and used for chemical yield monitoring. (**R10**)

4.4 Measurement methods for radionuclides

Different measurement methods are used depending on radionuclides. Liquid scintillation counting is used for the measurement of ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{63}Ni, and ^{129}I. Beta counter (Ultra low background gas flow GM counter) is used for ^{90}Y(^{90}Sr) and ^{99}Tc. Alpha spectrometry is used for ^{241}Am, ^{244}Cm, ^{238}Pu and $^{239,240}Pu$, ICP-MS for ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{99}Tc. Gamma spectrometry is

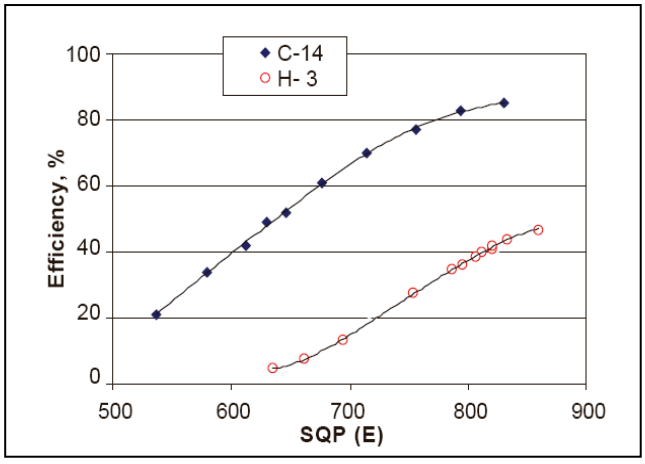
used for ^{94}Nb. Radioactive tracers (^{85}Sr and ^{99m}Tc) are used to monitor chemical yields. ICP-OES is used for measurement of stable elements applied for chemical yield measurement. These methods are briefly described below.

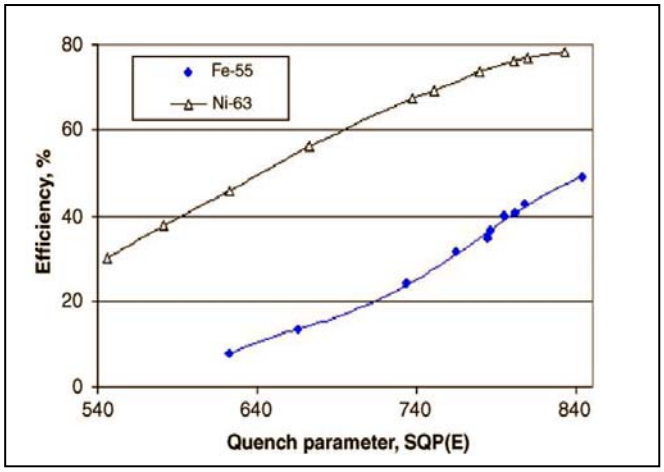
4.4.1 Liquid scintillation counting (LSC)

LSC is used for the measurement of radionuclides (including ^{3}H, ^{14}C, ^{36}Cl, ^{63}Ni, ^{90}Sr and ^{99}Tc, ^{129}I) decaying by emission of beta particles, whereas ^{41}Ca, ^{55}Fe decaying by electron capture are measured through Auger electrons.

Because the beta spectrum is continuous with energy distribution from zero to the maximum energy of the beta particles, the spectra of all beta emitters overlap with each other. Therefore, each beta-emitting radionuclide has to be separated from all others to avoid spectrum interferences. This is achieved by chemical separation as described above. Due to quench effects in the LSC, especially for measurement of radionuclides emitting low-energy beta particles and Auger electrons, the final sample solution has to be converted or conditioned in a less quench medium to reduce the quench effect, and quench curves of individual radionuclides have to be made using standard solutions in a medium similar to that of the final separated sample. Fig. 3 shows the quench curves of tritium, ^{14}C, ^{41}Ca, ^{55}Fe and ^{63}Ni measured using standard solution of the individual radionuclide and the external quench parameter (SQP(E)) feature in QuantulusTM 1220 LSC. Various approaches are applied for different radionuclides to reduce the quench effect. For ^{3}H measurement, water samples are filtered and distilled to remove particles and chemical quench reagents. Solid samples are decomposed using the combustion method and tritium is converted to tritiated water for measurement. For the measurement of ^{14}C in water, the samples are first pre-concentrated by evaporation. ^{14}C is separated by combustion of the evaporated water or solid samples to convert ^{14}C to CO_2, which is trapped in an amine solution for measurement. For the measurement of ^{36}Cl, chlorine is separated from the samples, and converted to NH_4Cl form and prepared in NH_4NO_3 solution to get rid of the high quench effect caused by AgCl precipitate or concentrated NH_3 medium in the traditional method for ^{36}Cl. This makes the counting efficiency as high as 95% for ^{36}Cl in our method. For the determination of ^{41}Ca by counting Auger electrons (<3.6 keV) using LSC, separated Ca as $Ca(OH)_2$ is dissolved in HCl and conditioned to a neutral pH. In this case the counting efficiency of ^{41}Ca can be improved to be higher than 15%. For the measurement of ^{55}Fe by counting Auger electrons (5-6 keV) using LSC, the colour quench from the brown colour of

Fe^{3+} solution is very serious, which significantly reduces the counting efficiency to be as low as about 3%. A novel method is developed to eliminate the colour of Fe^{3+} by using H_3PO_4 to dissolve the $Fe(OH)_3$ and convert Fe^{3+} to colourless complex with H_3PO_4. This approach improves the counting efficiency of ^{55}Fe to be higher than 35%. The quench effect for high-energy beta emitters such as ^{63}Ni, ^{90}Sr (^{90}Y), ^{99}Tc, and ^{129}I is normally less serious, but it is still needed to convert the medium of the separated solution to neutral and completely remove the high quench chemicals, such as CCl_4. The prepared solution is finally mixed with a scintillation cocktail for the measurement. Different cocktails are chosen to optimize measurements. For most of radionuclides, Ultima Gold LLT scintillation cocktail (PerkElmer Inc.) is used. This cocktail can be mixed with a high volume and salt concentration solution, which improves the figure of merit. For the measurement of ^{14}C, the separated ^{14}C in CarboSorb is mixed with Permafluor E^+ scintillation cocktail. It was observed that ^{14}C in carbonate form is not stable in other scintillation cocktail such as Ultima Gold, causing a variable counting efficiency of ^{14}C. For the measurement of ^{3}H when using the combustion method, Monophase S scintillation cocktail is used.





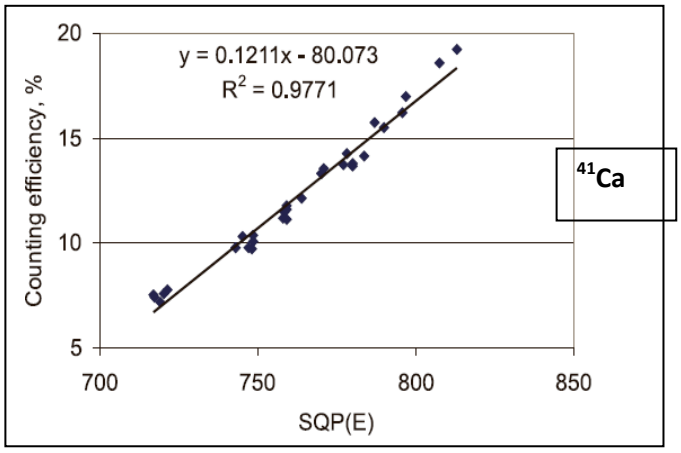


Fig. 3 Quench curves of LSC for ^{3}H, ^{14}C, ^{55}Fe, ^{63}Ni and ^{41}Ca

Besides the quench curves, a standard is always measured together with samples for quality control. In addition, process blanks are prepared using the same procedure as for the samples but without sample. These blanks are measured with the same batch samples representing contributions from chemicals used in the separation as well as instrument background. The blank signal is subtracted from the signal in the samples to get a net signal of samples.

The measured results as count rate (CPM) with the measurement parameters including quench parameters are recorded and stored in Excel files for further analysis.

For beta measurements, the radioactive tracers are removed before measurement either by decay (^{99m}Tc) or by chemical procedures (^{85}Sr). Stable isotopes are used for chemical yield monitoring of ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{63}Ni, and ^{129}I, which is measured by ICP-OES or ICP-MS. For tritium and ^{14}C, no chemical yield tracers are used, but experiments have confirmed quantitative recovery of tritium and ^{14}C in the combustion method. Since no chemical separation procedure is involved in the determination of tritium in water, no chemical yield tracer is needed.

4.4.2 Alpha spectrometry

Alpha spectrometry is used for measurement of alpha emitters, including ^{238}Pu, ^{239,240}Pu, ^{241}Am and ^{244}Cm. The separated radionuclides are electrodeposited on stainless steel discs for counting. ^{242}Pu and ^{243}Am are added in the beginning of the chemical separation to monitor the chemical yield of Pu isotopes and ^{241}Am and ^{244}Cm, respectively. Due to similar chemical properties of Am and Cm, ^{243}Am can be used for both radionuclides. Because the counting efficiency in the alpha spectrometry does not vary with energy of alpha particles, the activity of the target radionuclides can be calculated directly from the activities of the added ^{242}Pu and ^{243}Am tracers and the measured counts of target radionuclides and tracers. The process blanks are always prepared using the same methods as the samples, and measured by the same alpha detector. The blank results are subtracted from those of the samples during activity calculation.

4.4.3 Beta counting

Low-level gas flow GM counters (Risø GM Multicounter system) are used for the determination of ^{90}Y(^{90}Sr) and ^{99}Tc. Due to its extremely low background (<0.2 CPM) and relatively high counting

efficiency (about 54% for ^{90}Y and 44% for ^{99}Tc), the detection limit of this method is better than that of LSC.

For this measurement, the separated sample is prepared on a disc and placed in a holder. For ^{99}Tc, the separated ^{99}Tc is electrodeposited on a stainless steel disc and mounted in a holder using thin Mylar film. For $^{90}Y(^{90}Sr)$ measurement, ^{90}Y is finally precipitated as $Y_2(C_2O_4)_3$, which is filtered through a small filter (15mm in diameter), and mounted in a holder using Mylar film. The process blanks are also prepared and measured together with samples, and the contribution from the blank is subtracted from the sample signals. Standards of individual radionuclides in the same geometry and matrix as the samples are used to calibrate the detectors for counting efficiency.

4.4.4 Inductively coupled plasma mass spectrometry (ICP-MS)

ICP-MS is used for the determination of ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{99}Tc. In addition, it is also used for some stable elements using as carrier/tracer such as ^{127}I or ^{36}Cl for chemical yield monitoring. A Thermal X series ^{II} ICP-MS (Thermal Electron Corporation), a quadrupole type mass spectrometer, installed in our group at Risø-DTU, is used for this purpose.

The sample is normally prepared in 0.5M HNO_3 solution, except for iodine and chlorine which need to be analyzed in ammonium solution. Internal standard such as In^{3+} for Pu, Re for ^{99}Tc, or Cs^+ for iodine and chlorine, is added to the solution for monitoring introduction efficiency. The standards of individual radionuclides or stable isotopes are prepared and measured with samples in the same instrumental condition for instrumental calibration. The process blanks are prepared using the same procedure as samples and measured with samples. The blank signals are subtracted from the signals of the samples when the measurement results are calculated.

5. Progress on development and optimization of analytical methods for decommissioning

5.1 Analysis of PCB paint materials from hot cell at Risø, Denmark

The decommissioning of the nuclear facilities at Risø, Denmark, involves decontamination of 6 hot cells. During operation from 1964 to 1989 the hot cells were used for examination of spent fuel elements and production of irradiated cobalt pellets. Thus the cells are contaminated with actinides,

fission products and some activation products. The interior compartments of the concrete construction are sealed with a steel liner, which is coated with epoxy paint. The paint contains significant levels of PCBs and lead. The decontamination of the inner side of the hot cell is going to be carried out by sandblasting the paint off the steel liner. It is estimated to get app. 100 barrels, or 10,000 liters, of sandblasting material mixed with contaminated paint particles. A subsample is collected from each barrel, which is going to be analyzed to estimate the total inventory of the radionuclides.

The challenge is to establish a method for analyzing the alpha emitting radionuclides. In general, this analysis requires fully decomposing the materials to release the radionuclides to a solution from the matrix. Some preliminary testing has been performed. It was found that the epoxy paint (PCBs) cannot be decomposed by conventional methods, e.g. acid digestion using aqua regia and ashing at less than 1000 °C. A leaching method using hot *aqua regia* was investigated to release radionculides from the materials. Due to the relatively high concentrations, low energy gamma and X-ray spectrometry was used to estimate the leaching rate of actinides. It was found that most of Pu and Am in the materials can be released with acid leaching, while part of Cs was released from the materials. Combined with chemical separation as described in this report, the determination of isotopes, Am, Np and Cm in this kind of waste can be carried out. A detailed analysis of the leaching of the individual samples is going on. The results will be presented with a poster at the NKS seminar in Decommissioning of nuclear facilities, Studsvik, Nyköping, Sweden, 14 – 16 September 2010

5.2 Analysis of concrete for free and bound tritium and 14C.

In bio-shielding concrete in the nuclear reactor, tritium may exist in two forms, tritiated water which is produced in the heavy water as well as in the reactor and adsorbed in the concrete or produced in the concrete, and bound tritium which is mainly produced in the concrete by activation of Li in the concrete with neutrons from the reactor. Since the property of these two forms of tritium are significantly different, the tritiated water are high mobile and is easily released to the air when is exposed to the atmosphere, while the bound tritium is more stable in the concrete. This is very important for the depository of this kind of waste.

In DR3 reactor at Risø, due to the leakage of reactor tank, heavy water was leaked and adsorbed in the underneath concrete, for estimation of tritium species and inventory in these concrete, a method was developed to analyze the free tritium and bound tritium in concrete. A concrete core was drilled under

freezing; the collected concrete was immediately frozen by liquid nitrogen, and sealed in a plastic bag which is sealed in a plastic bottle. Before samples preparation and analysis, the samples was taken from the bottle and frozen again in liquid nitrogen.

The frozen concrete was crushed in in the glove box, 1-4 g crushed sample was transferred to a three-neck flask, which was also frozen in liquid nitrogen. The wet sample and flask was weighted, the flask was connected to the heating system, the compressed air was connected to the system to blow the sample and transfer the evaporated water vapor and CO_2 from the sample through the system. The sample was then heated to 150-200 °C for 1 hour. The system was then dismantled. The condenser, collector and connections were washed with water, and washes were combined with the trap water in the first bottle, the water in total was weighted, and then used for the free tritium measurement. The dried sample and flask was weight, the dried sample in the flask was then transferred to a vial for further analysis for bound tritium. The empty flask was weighted. The wet weight and dried weight of the sample was then calculated. The 10 ml of tritium trap solution (water) to a vial, 10ml of Ultima Gold LLT cocktail was added and mixed for the measurement of tritium.

For the determination of bound tritium and ^{14}C, 2 aliquots of about 0.3 g dried sample of each was weight to a cellulous pad, about 0.3 mg of cellulous powder was added and mixed with concrete sample. The samples were then decomposed using Sample Oxidizer at 1100 °C to separate bound tritium and 14C. The separated tritium and C-14 were measured using LSC. Table 4 given analytical results of sample samples in the concrete core from DR3.

Table 4 Free and bound tritium and ^{14}C in concrete from DR3 reactor at Risø, Denmark

Sample ID	Loss of weight at 150°C, %	Free Tritium in concrete, Bq/g	Bound tritium in concrete, Bq/g	Bound ^{14}C in concrete, Bq/g
T5-A2	1.838	9.08	0.65	2.04
V1-O5	2.274	329	12.95	25.80
V3-8	2.808	17.0	0.31	14.10
V3-14	2.176	38.1	1.44	3.73
V5O	4.609	81000	119.47	0.27
V7O	4.461	2570	37.71	0.71

5.3 Determination of 45Ca and 63Ni in reactor concrete

In Helsinki University, reactor concrete has been analyzed to determine ^{45}Ca for the estimation of ^{41}Ca in the concrete, to determine ^{63}Ni and gamma-emitting radionuclides (Ervanne et al. 2009). ^{41}Ca is a long-lived neutron activation product (half-life 103 000 years). Instead of measuring ^{41}Ca, ^{45}Ca was measured and the activity concentration of ^{41}Ca was calculated based on the determined ^{45}Ca activity. Concrete samples were not completely dissolved. Instead calcium was partially leached from concrete and the total calcium content in the concrete was determined by nondestructive XRF-analysis. Calcium was separated from the leachates using cycles of precipitation and ion exchange. In addition to ^{45}Ca, seventeen γ-emitting radionuclides were detected directly in the concrete samples by γ-spectrometry. Ratios of ^{45}Ca activity concentrations to those of γ-emitting nuclides in the concrete samples with varying dose were more or less identical. Seven concrete samples from the Loviisa NPP irradiated for one year at the inner surface of the reactor concrete shield. The samples were exposed to varying (unknown) neutron fluxes. Gamma-emitting radionuclides were determined by gamma spectrometry. Gamma-emitting radionuclides were determined by gamma spectrometry. Calcium-45 was determined by liquid scintillation counting after radiochemical separation. Stable calcium in the concrete was determined by XRF and Ca in the acid leachate by AAS. ICP-MS was used to determine the elemental composition of the concrete. Fig. 4 shows a separation procedure for this analysis.

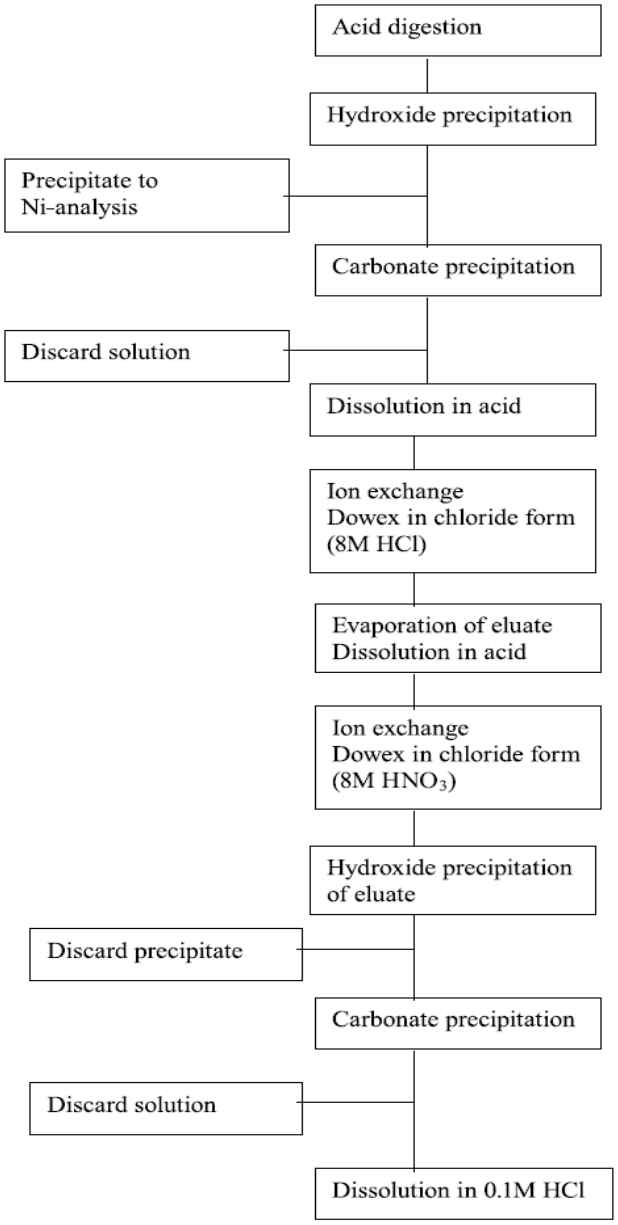


Fig. 4 Radiochemcial procedure for separation of ^{45}Ca from reactor concrete

5.4 Analysis of bullet concrete for tritium and 14C

A special concrete was used as part of the biological shield in DR-3 reactor, which is made of 87% of iron balls and 13% of ordinary concrete. The iron ball was supposed to act as a shielding for radiation from the reactor. Since the concrete was exposed to tritiated water in the irradiated heavy water, a tritium in the concrete was expected. A method was developed to determine tritium in this material. Some concrete was taken from different part of concrete stringers from the heavy water room, which was then crushed; the concrete was then separated from iron ball by removing the attached concrete from the iron ball. The concrete part was then ground to fine powder. About 0.1 g of concrete powder was mixed with cellulous powder in a cellulous cone, 3-4 pieces of iron ball was mixed with cellulous powder in another pad. The tritium in both concrete part and iron ball was separated by combustion using Sample Oxidizer, and measured using LSC. Table 5 show the analytical results of some of this material. It can be seen that most of tritium is bound to concrete part, while the tritium in iron ball is only a small fraction. By taking representative samples, the inventory of tritium in these concrete (about 10 tones) was estimated.

Table 4 Analytical results of tritium and ^{14}C in concrete samples from DR3 heavy water room

Sample I.D	Sample	^{3}H concentration, Bq/g	^{14}C concentration, Bq/g
S1A	iron ball	19.3±4.5	0.17±0.06
SIA	inner concrete	3828.6±231.2	7.55±1.29
S1C	iron ball	66.1±10.3	0.22±0.12
SIC	inner concrete	4831.2±1083.3	3.92±0.68
S1B	iron ball	56.7±25.0	0.15±0.04
SIB	concrete	7925.9±891.0	1.68±0.20

5.5 Automated and rapid analytical method for 239Pu, 240Pu and 237Np.

Determination of transuranics, especially the alpha emitting transuranics, a tedious chemical separation has to be applied to separate the target radionculides from the matrix and interfering isotopes. The conventional separation method normally takes a few days to one week for one batch of samples. This

is not suitable for the analysis of a large number of samples required in the characterization of decommissioning waste.

At Risø-DTU, an automated analytical method for rapid and simultaneous determination of plutonium and neptunium has been developed. A chromatographic column packed with AG MP-1M macro-porous anion exchanger or extraction chromatographic resin was incorporated in a sequential injection (SI) system to implement the automated separation of plutonium along with neptunium from the matrix elements and interferences. The sorption and elution behaviour of plutonium and neptunium onto chromatographic column was investigated. The results revealed the pore size was a critical factor to actualize the similar separation behaviour of plutonium and neptunium on the anion exchanger. By this means, the total on-column separation could be completed within 3.5 hours, more importantly, which significantly improves the sample throughput and enables to analyses large number of samples in a short time. Fig. 5 illustrates the principle of the sequential inject set up for automated and rapid separation of Pu and Np from samples.

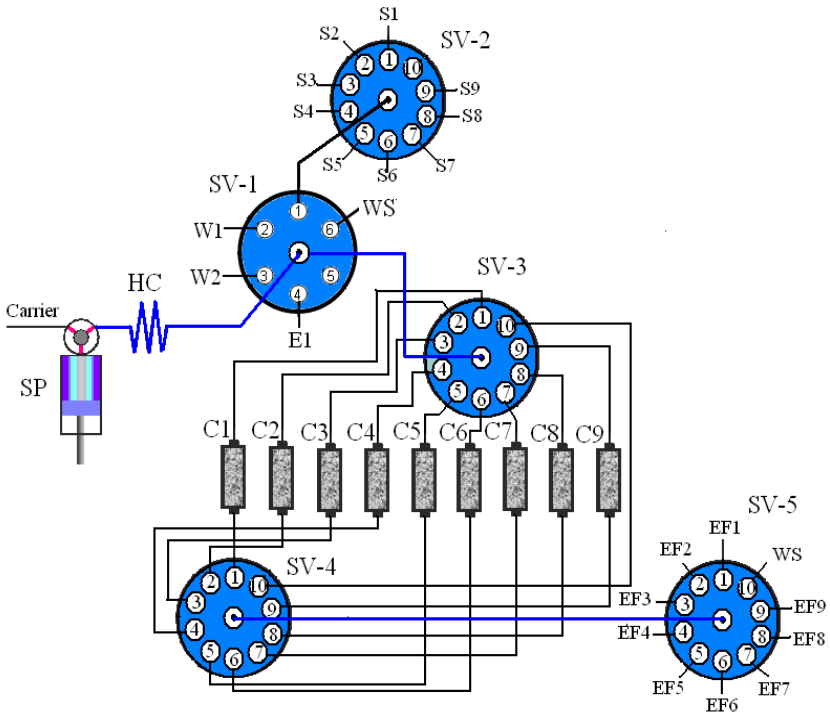


Fig.5. Schematic diagram of the SI system for automated determination of Pu in environmental solids W1: 8 mol/L HNO_3; W2: 9 mol/L HCl; E1: 0.5 mol/L HCl; WS: waste; S1-S9: ports for sample loading; EF1-EF9: ports for eluate collection; SP: syringe pump; SV-1 - SV-5: selection valves; HC: holding coil; C1-C9: nine exchange columns.

5.6 Estimation of radioactivity in Barsebäck NPP by measurement and calculation

The two units in Barsebäck NPP have been closed in November 1999 (B1) and May 2005, and will be decommissioned in about 10 years. Some work has been carried out to estimate the activity inventory in the power plant, including measurement and calculation.

Two concrete cores with length of 150 cm, and diameter of 45 mm have been taken from the middle of the side of the reactor core and 1.3 m below reactor core bottom. The concrete cores were sliced into pieces of 30-50 mm length in order to quantify the radial distribution of induced activity. The sliced samples were analyzed for gamma emitters by gamma spectroscopy including ^{40}K, ^{58}Co, ^{60}Co, ^{152}Eu, ^{154}Eu and ^{134}Cs, while beta and alpha emitters such as ^{3}H, ^{14}C, ^{63}Ni, ^{55}Fe and transuranics were not analyzed. The chemical composition of samples from the reinforcement bars in the concrete sample was also analyzed, which is used as input for the modeling calculation of the distribution of radionuclides in the concrete core. Activity in isolation material (Caposil) just outside RPV and an Al-plate (part of the isolation) were also determined. Fig. 6 shows the distribution of ^{60}Co and ^{152}Eu in two concrete cores. Fig. 7 compares the measured activity of ^{60}Co and ^{152}Eu with the model calculation.

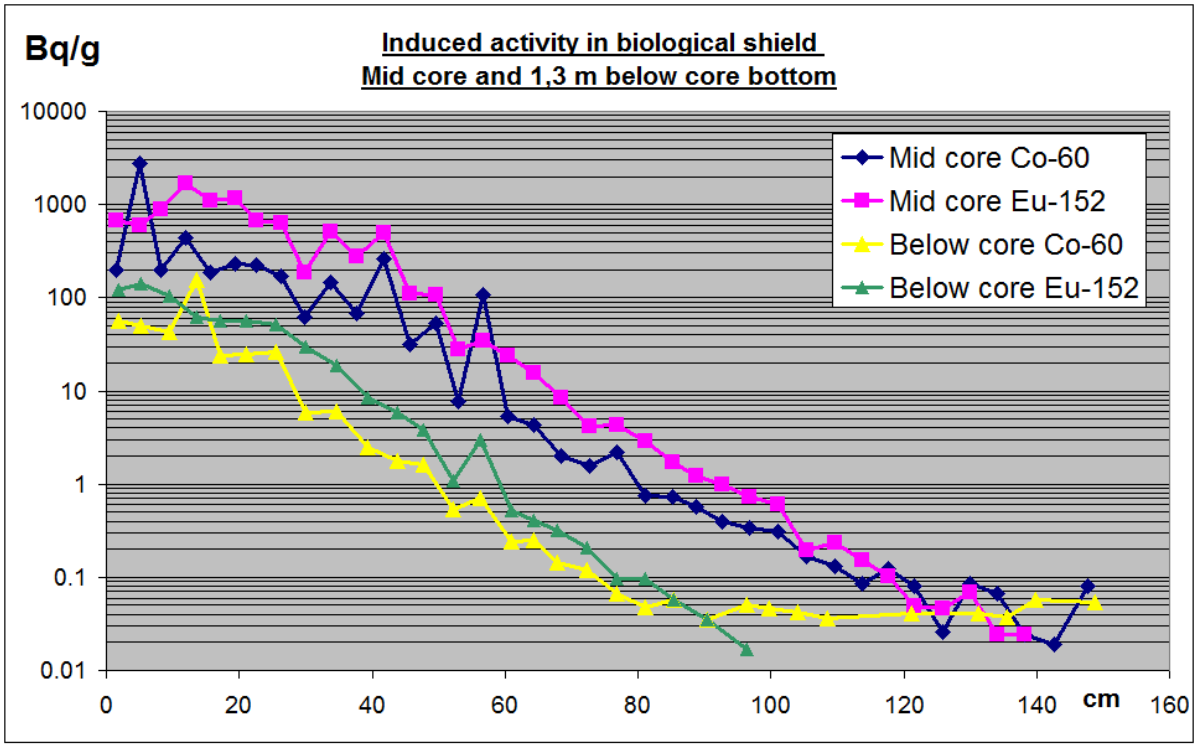


Fig. 6 Measured ^{60}Co and ^{152}Eu distribution in two concrete core from the Barsebäck reactor B1.

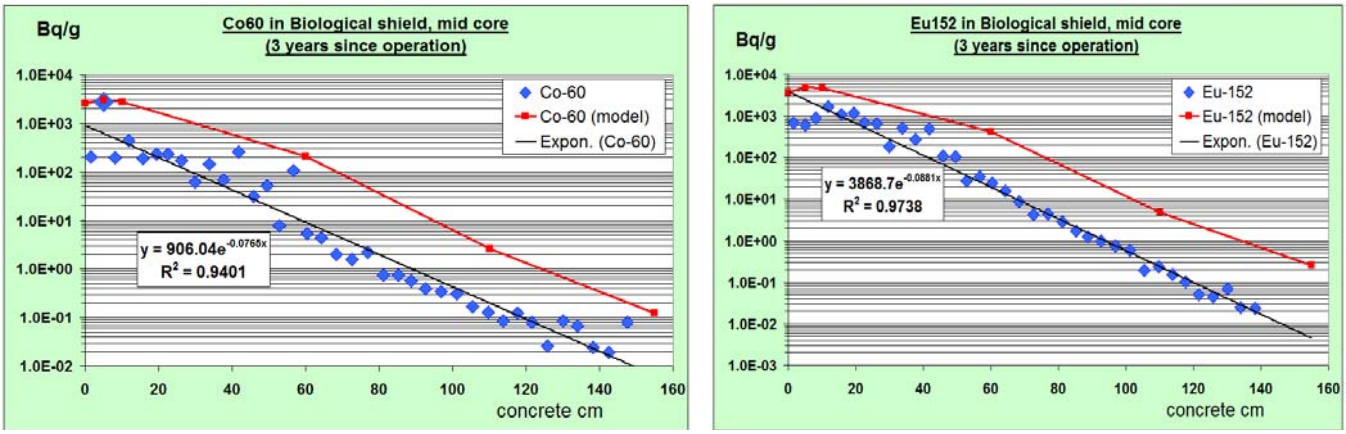


Fig. 7. Comparison of the measured activities of ^{60}Co and ^{152}Eu in the concrete cores with the model calculation

It was observed that ^{60}Co and ^{152}Eu are the dominating gamma radionuclides in the bio-shielding concrete, and the activity has an exponential attenuation to distance from reactor tank; fragments of reinforcement bars present in the part of the sample tube closest to the reactor tank, shows the highest concentration of ^{60}Co activity; the model overestimate the activity by a factor of 10-50. The relative difference increases with distance from reactor tank. Calculations show that induced activity varies by a factor of 10 with angel. Samples were taken at 335^{o}. This angel represents a mean value of activity concentration. Chemical compositions of the concrete have an influence on the activity concentration. Not only the target nuclides, but also the concentration of hydrogen and boron has an major impact on the neutron flux. The level 0.1 Bq/g correspond to the distance of approximate 120 cm to reactor core. As the outer part of the biological shield also contains reinforcement bars with higher concentration of ^{60}Co, 100 % of the biological shield in core region must be regarded as radiological waste. The activity concentration in the sample 1.3 m below lower core level, is 1/10 of the activity concentration in sample from mid core region. The level 0.1 Bq/g corresponds to the distance 100 cm concrete for this "below core sample.

One concrete core samples were taken from bottom of floor drainage cavity of the main circulation pump in containment to investigate the penetration of radionculides into highly contaminated epoxy covered concrete surface. Fig. 8 shows the distribution of ^{60}Co and ^{137}Cs in these samples.

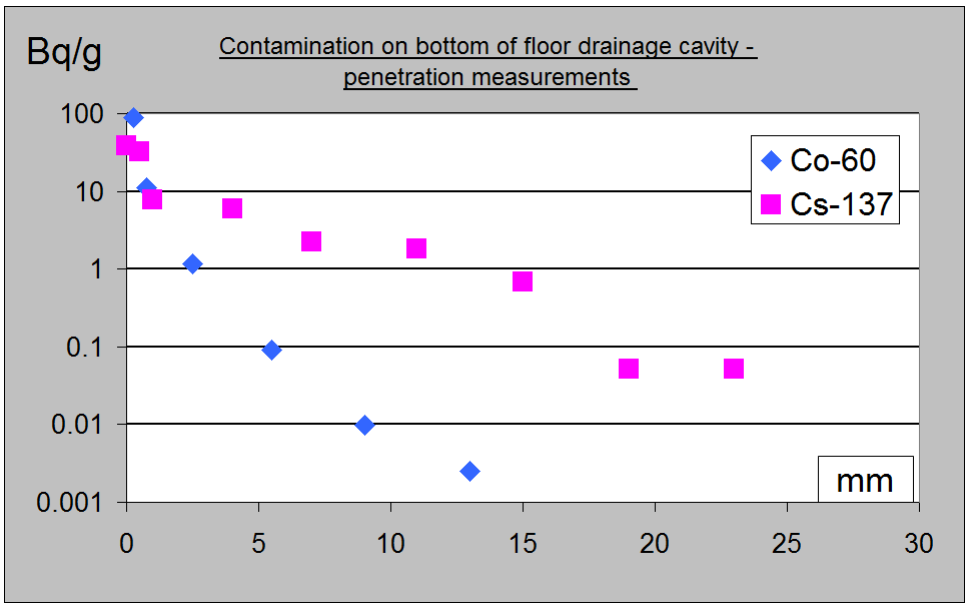


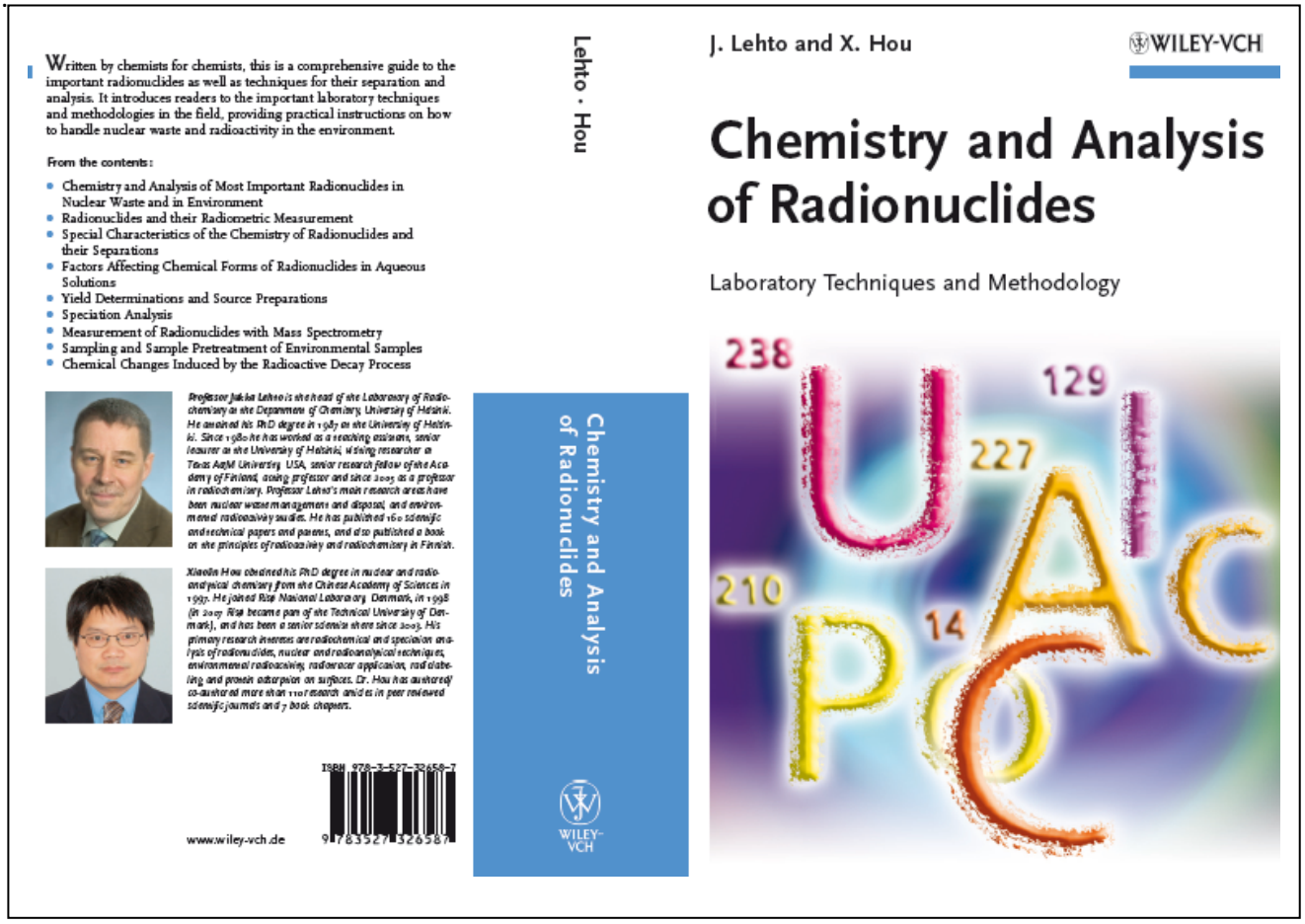
Fig. 8 Distribution of radionuclides through epoxy covered concrete

The results indicate that ^{60}Co and ^{137}CsCs were detected and both showed an exponential attenuation with depth. ^{60}Co dominates the surface contamination, but the penetration was insignificant. ^{137}Cs defines the contamination depth to 12 mm overall.

6. A book on "Chemistry and Analysis of Radionuclides"

In the project proposal, a review article was planed to review and compare the available radiochemical analytical methods of decommissioning waste. In practice, this was replaced by a book entitled on "Chemistry and Analysis of Radionculides, which is authored by the project partners Jukka Lehto in University of Helsinki and Xiaolin Hou in Risø-DT to be publication by Wiley-VCH in 2010, the book is now in press. The book consists of 19 chapters including 1) Radionuclides and their radiometric measurements; 2) Special characteristics of the chemistry and analysis of radionuclides; 3) Factors affecting chemical forms of radionuclides in aqueous solution; 4) Radiochemical separation methods; 5) Yield determinations and source preparations in radiochemical analyses; 6) Radiochemistry of alkali metals; 7) Radiochemistry of alkaline earth metals; 8) Radiochemistry of 3d transition metals; 9) Radiochemistry 4d transition metals; 10) Radiochemistry of lanthanides; 11) Radiochemistry of halogenides; 12) Radiochemistry of noble gases; 13) Radiochemistry of tritium and ^{14}C; 14)

Radiochemistry of lead and polonium; 15) Radiochemistry of actinides; 16) Speciation of radionuclides; 17) Measurement of radionuclides with mass spectrometry; 18) Sampling and pre-treatment of environmental samples; 19) Chemical changes induced by radioactive decay. The cover of the book is shown below.



7. Conclusion and perspectives

By this project, a preliminary Nordic network on radioanalysis of waste samples from decommissioning and waste depository activities has been established. Two project meetings and one NKS workshop with cooperation with another NKS project RadWorkshop, have been organized to promptly discuss the problems and exchange the experience. The radioanalytical methods developed and applied in the Nordic labs as well as worldwide were reviewed, a book on this issue has been written by the project partners to be published in 2010, a brief description of these methods are

presented in this report; By involving the nuclear industries in this project, the present requirement from the Nordic decommissioning activities and the present problems in the radiochemical analysis of nuclear waste were identified; Among project partners, some development and optimization of radioanalytical methods for decommissioning waste have been carried out, the progress in this issue are presented in this report. During the project meeting, the project partners visited the Barsebäck NPP and radiochemical laboratory at Risø-DTU, and a demonstration of analytical procedure for some radionculides in nuclear waste in Risø-DTU were organized during workshop.

A close collaboration among project partners were discussed and expected to be carried out in the future, some activities, such as validation of the model estimation of ^{36}Cl and ^{129}I in operation waste from Swedish light water reactors by analyzing the real samples for ^{129}I and ^{36}Cl using the methods developed at Risø-DTU, and investigation on the possibility to make the developed models as a benchmark for the estimation of ^{129}I and ^{36}Cl in the operation waste from Swedish power reactors, and development of method for the determination of uncommon radionculides in waste samples such as ^{93}Mo, ^{94}Nb and ^{93}Zr are some examples to be involved in these collaboration. Such collaboration will be benefit to both Nordic nuclear industries and radiochemical laboratories in Nordic countries.

All partners agreed that the established network is very useful not only in the exchange of knowledge and experience, but also in the improvement of the collaboration in this field, and this network should be continued and enlarged to involve more labs/industries in Nordic countries in the next step. To improve this network, a website is expected to be established in the near future acting as a platform to exchange the ideas, experiences, as well as requiring answer for questions.

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Abstract

The NKS-B RadWaste project was launched from June 2009. The on-going decommisioning activities in Nordic coutries and current requirements and problems on the radiochemical analysis of decommissioning waste were discussed and overviewed. The radiochemical analytical methods used for determination of various radionculides in nuclear waste are reviewed, a book was written by the project partners Jukka Lehto and Xiaolin Hou on the chemistry and analysis of radionuclide to be published in 2010. A summary of the methods developed in Nordic laboratories is described in this report. The progresses on the development and optimization of analytical method in the Nordic labs under this project are presented.

Key words Radioanalysis, radionculides, decommissioning, radioactive waste

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